

# Investigation of Carbon-based Aluminum Thermal Management Composites Sensitized by High-pressure Impregnation Method

Nan Jiang\*, Jamie Novak

Applied Nanotech, Inc., 3006 Longhorn Blvd., Suite 107, Austin, USA

njiang@appliednanotech.net

**Abstract-** Power electronic packaging highly demands the innovative light-weight heat dissipation materials with a combination of the low coefficient of thermal expansion (CTE) and the high thermal conductivity. In this study, the high-performance carbon based aluminium thermal management composite has been successfully developed by infiltrating porous carbon matrix with Al at high-pressure conditions. It is found that Al can be effectively injected into the matrix tiny pores as small as 40 – 50 nm, forming a smooth C/Al interfacial contact. The C-Al composite presents attractive thermal behaviours including high thermal conductivity (over 400 W/m-K), and a low CTE value that is smaller than 7 ppm/ K and matches well with conventional semiconductors. Also, C-Al composite is a light-weight material which has a density only about 2.3 g/cm<sup>3</sup>. In addition, the ceramic thin sheets can be tightly bonded to C-Al composite to form the dielectric composite plates using the same impregnation approach. The present C-Al composite is considered to be a promising candidate for a variety of thermal management applications in power electronic and photonic industrial fields.

**Keywords-** C-Al Composite; Thermal Conductivity; Low CTE; Porosity; High-Pressure Impregnation

## I. INTRODUCTION

Electronic and photonic devices have been pushed toward faster and higher power, with increasing demand for heat dissipation [1-3]. The average operating temperature of an electronic component is directly related to the reliability, efficiency, lifetime, and performance of the product, and over 55% of failures in electronic components are due to high operating temperatures. As the physical size of electronic components continue to shrink and become faster and more powerful, thermal management becomes more and more important. These systems require thermal management materials capable of effectively dissipating heat while maintaining physical compatibility with the package and die [4, 5]. New high performance and low cost materials are needed to address the significant deficiencies of thermal management materials today as well as for emerging applications in the future. The thermal management materials with a combination of high thermal conductivity and low CTE are highly demanded. Most traditional low CTE materials such as W/Cu, W/Mo have thermal conductivities that are no better than those of aluminium alloys, about 200 W/m-K [6, 7].

On the other hand, the outstanding thermal and mechanical properties of carbon allotropes have driven considerable interest in the development of novel thermal transfer materials. Worldwide development effort of nano-carbon heat sink composites started over a decade ago after the discovery of carbon nanotubes (CNT) and prediction of

their thermal conductivity properties. These novel carbon related forms include CNT, graphene, and nano-diamond particles enhanced composites [8–10]. These composite materials are expected to be superior over others with respect to CTE, thermal conductivity and density. Meanwhile, filling the pores of a carbon matrix with metals thereby forming carbon matrix composites has also been attempted to improve the characteristics of carbon materials. As-fabricated graphitic blocks are often highly porous limiting the material's continuity and degrading the material's capability. Filling the pores of graphitic materials with metals to form carbon based metal composites has been considered to be a promising research topic [11]. However, due to the poor affinity between carbon and metals, few examples showed sufficiently filled pores or the distinct performance improvement.

In this study, we present a unique carbon based metal composite obtained through pressure injection technology that consists of a carbonaceous matrix containing 20wt% aluminium dispersed metal component. It is a sophisticated composite with low thermal expansion, high thermal conductivity and excellent thermal diffusivity.

## II. EXPERIMENT

The carbonaceous matrix used for Al impregnation is produced by means of extruding a graphite/pitch mixture followed by high temperature sintering. The carbon matrix includes graphitic backbone and pores; the porosity is about 25%. A high-pressure impregnation process is used for injecting molten Al into the carbonaceous matrix. The temperature, pressure and impregnation time are optimized at about 730 °C, 100 MPa and 10 minutes, respectively. The material composition and microstructure are evaluated by transmission electron microscopy (TEM) and scanning electron microscopy (SEM) equipped with the dispersive x-ray spectroscopy (EDS). The thermal properties are investigated by laser flash measurement (KEM LFA-502), and a PoreMaster 60 porosimeter was employed to exams material porosity and pore size distribution before and after Al impregnation.

## III. RESULTS AND DISCUSSION

The material developed in this study begins with a porous graphitic carbon. The final form consists of carbonaceous matrix of which most of pores are filled by aluminium. Fig. 1 is a typical image showing C-Al composite appearance. The black contrast area is metric carbon, in which the white domains are aluminium-filled pores. The C-Al composite fabricated in this study is in a block shape and can have a maximum dimension of 250 mm x 200 mm x 150 mm. This kind of C-Al composite has a nice machinability, and

fabrication of complicated shape parts is feasible as revealed in the inset of Fig. 1, wherein C-Al composite has been processed into a fin-type heat sink parts.



Fig. 1 Images of C-Al composite and a machined heat sink parts (inset)

Table I outlines the major thermal properties of the C-Al composite. Using a carbon matrix as the majority component in the composite allows achieving a small CTE (no more than 7 ppm/K) providing compatibility with commonly used semiconductor materials such as Si, GaN and GaAs. The low CTE feature offers the opportunity for direct attachment of electronic components (even bare chips) on the composite material without cracking or delamination of the semiconductor components due to thermal expansion mismatch. It is feasible to manufacture the material with anisotropy in the alignment of the graphitic carbon structure.

TABLE I THERMAL PROPERTIES OF C-AL COMPOSITE

Orientation	Unit	X-Y (Normal to Extrusion Direction)	Z (Extrusion Direction)
Thermal Diffusivity	Cm <sup>2</sup> /Sec	1.25	2.55
Thermal Conductivity	W/m-K	200	425
CTE	Ppm/K	2.2	7
Specific Heat	J/Gk	0.75	0.75

For example, when the carbon starting material is extruded the C-Al composite has a thermal conductivity of 425 W/m-K in the extrusion direction (Z direction).

This compares favorably to bulk Al and Cu thermal conductivities of 237 W/m-K and 390 W/m-K, respectively. In general, a substance's thermal diffusivity is an important factor contributing to fast heat transfer and rapid temperature equilibrium, helping eliminate the "hot spots" generated in the electronic system. The Z direction thermal diffusivity is 2.55cm<sup>2</sup>/sec, which is about 2.5 – 3 times higher than that of Cu or Al. Although the composite is anisotropic, their thermal diffusivity is still higher than that of Al and Cu along X-Y directions (normal to the extrusion direction), and the thermal conductivity of X-Y directions can also be as high as those of Al alloys. Another advantage of using carbon as a base material is the relatively low mass density (2.3 g/cm<sup>3</sup>) which is lighter than Al ( $\rho = 2.7 \text{ g/cm}^3$ ). Low density is an important consideration [12] in aerospace and automotive applications where saving just a few grams can be the only route to maximize the performance of the overall system. The combination of the improved thermal performance and lighter

weight for a C-Al composite material is advantageous for a wide variety of thermal management applications.

Thermal management composites must rely on several factors to achieve good thermal conduction, including: (i) high thermal conductivity of the parent materials; (ii) lack of voids and cavities in the material and (iii) in many cases, creation of well contacted interface between the two parent materials. The C-Al composite has the potential to achieve excellent thermal conductivity since both aluminium and graphitic carbon have high thermal conductivity values. However, the last two criteria offer much room for improvement.

In order to well understand the nature of the material's outstanding thermal performance, C-Al composite's microstructure and C/Al interface have been investigated. Fig. 2 shows a SEM image (upper), and the corresponding EDS mappings of carbon element (middle) and aluminium element (lower) taken from the C-Al composite. Fig. 2 reveals that Al (as indicated by arrows) is infused into the irregular shape pores with few voids visible. This point has also been evidenced by EDS mapping pictures, wherein the contours of Al and C regions exactly match with each other. These images tell for sure that Al has been impregnated into carbonaceous matrix very well; and it fills pores and perfectly integrates with surrounding carbon matrix.

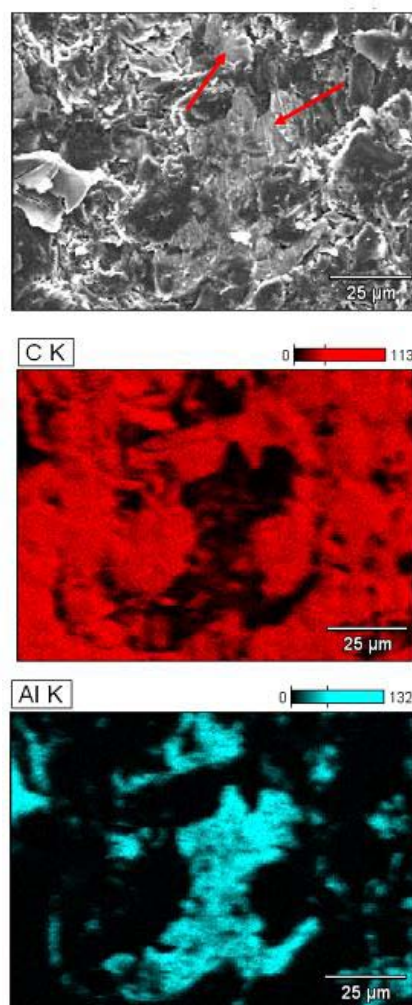


Fig. 2 A typical SEM image of the C-Al composite (upper) and the corresponding EDS mappings of carbon (middle) and aluminium (lower)

Material's porosity and pore size distribution before and after Al impregnation were studied using a PoreMaster 60 porosimeter system which allows measuring a wide range pore size distribution from nanometer up to sub-millimetre. The graphitic carbon matrix before Al impregnation has a porosity of about 25 vol. %, and after impregnation the total pore volume has been greatly reduced. The formed C-Al composite has porosity of only about 8 vol. %, namely near 70% initial pores are filled by Al. As the result, the material density increases from the  $1.75 \text{ g/cm}^3$  to  $2.30 \text{ g/cm}^3$ , and the C-Al composite consists of about 80 wt% of carbon and 20 wt % of Al. Comparing Fig. 3a and 3b, it is not difficult to find Al impregnation killed most pores that are greater 40 - 50 nm, but for the pores smaller than 20 - 30 nm, changes are minor, meaning the filling effectiveness for this impregnation process can extend to 40 - 50 nm tiny pores. We call this nanoscale filling as "nano-infiltration".

As mentioned above, it is well known for composite material synthesis, control over the interface between the matrix and the foreign fillers are especially important and represent one of the key factors affecting the final material's properties. In general, carbon materials and molten metal have poor wettability and affinity to one another. This creates a situation where the molten alloy does not want to wet the surface of the carbon. In other words, the high surface tension of the metal results in insufficient filling of the porous graphitic materials. This results in loss of contact between the two materials leaving voids and poor thermal performance. Careful control over the interface between the aluminium and carbon and the specify process parameters can overcome these limitations.

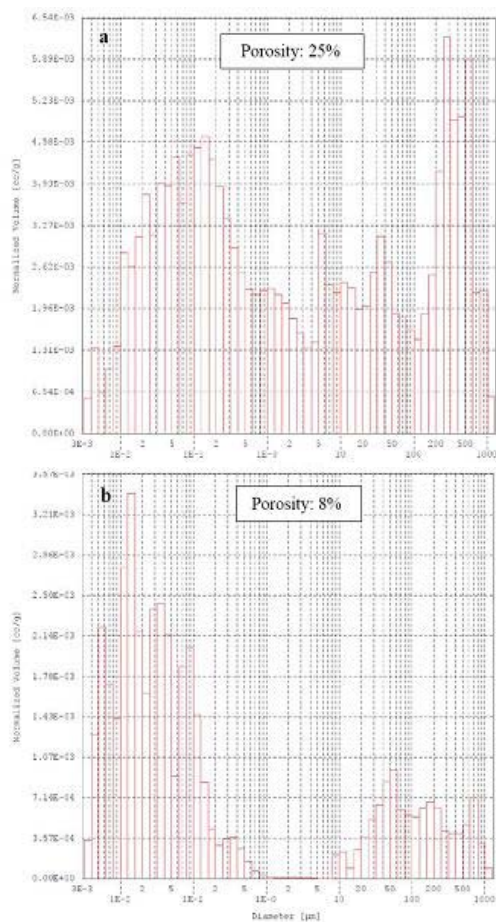


Fig. 3 The pore size distribution before (a) and after (b) Al impregnation

As one can observe from Fig. 4, the high-pressure impregnation allows aluminium to create a well matched interface at the nano-scale dimensions including contact angle and flatness, where the size of interfacial voids or slits (as noted by arrows) has been successfully suppress to less than 100 nm forming a closely contacted interface between the carbon phase and Al phase that ensures the heat flow can be smoothly conveyed from one phase to another phase. An organic combination of smooth interface contact, nano-infiltration, and the intrinsic advantages of Al and graphitic carbon is just the reason accounting for C-Al composite's excellent thermal behaviours.

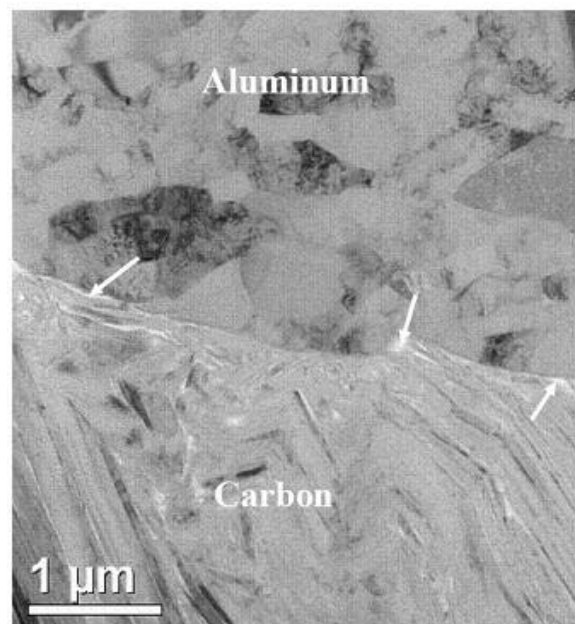


Fig. 4 A typical TEM image showing the C/Al interface  
The interfacial nano-voids and slits are indicated by arrows

In this study, to further extend the composites' application, we also developed the electrically insulating C-Al plate by attaching a ceramic sheet to the outer surface of a thermal management composite plate. In the areas such as IGBT (insulated gate bipolar transistor) or CPV (concentrating photovoltaic) cells, the electrically insulating materials with exceptional thermal performance are required. One normal way to provide electrical isolation is to adhere ceramic insulators onto traditional thermal management material surfaces using adhesives such as epoxy. However, the low thermal conductivity of epoxy can significantly raise the cross-sectional thermal resistance and impede the heat transport. In this study, we present a novel approach by which the insulating ceramic sheet can be directly bonded to the composite surface during the high pressure impregnation manufacturing process. This results in an electrically insulating material without using organic adhesives that could potentially reduce thermal performance. A carbon matrix plate and a ceramic sheet were previously stacked together, and during the high pressure impregnation process, as the molten Al is injected into the porous carbon to form composite, a part of molten Al can also be injected into the gap between that carbon plate and ceramic sheet, tightly connecting the two parts into one body as revealed in Fig. 5, where a silicon nitride sheet is bonded on C-Al composite. The combination of these materials provides a facile route to excellent adhesion. One can expect outstanding cross-sectional heat transport behaviours due to eliminating the

organic adhesives and even better thermal performances by further thinning the ceramic sheet thickness.

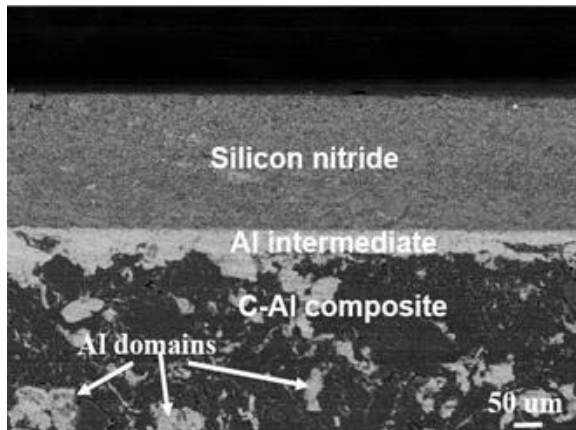


Fig. 5 SEM image of the C-Al composite with a silicon nitride dielectric sheet bonded on the top

#### IV. CONCLUSIONS

The C-Al thermal management composite material has been developed by means of high-pressure impregnation process.

At the optimized process conditions, molten Al can be effectively injected into the pores that can be as small as 40 – 50 nm in the carbon matrix creating a close contact at C/Al interface.

After impregnation, near 70% initial pores are occupied by Al fillers. The C-Al composite presents a low CTE, a high thermal conductivity and an excellent thermal diffusivity. Smooth interface contact, nano-infiltration, and the intrinsic advantages of Al and graphitic carbon are considered to be the three key factors accounting for C-Al composite's excellent thermal behaviours. The C-Al composite plates with the surface electrically insulating ceramic sheets have also been successfully produced by the same impregnation approach without using any foreign adhesive materials.

#### REFERENCES

- [1] Luedtke A., 2004, *Adv. Eng. Mater.*, 6, 142.
- [2] Mahajan R., Chiu C. P., and Chrysler G., 2006, *Proc. IEEE*, 94, 1476.
- [3] Lin W., Zhang R., Moon K. S., and Wong C. P., 2010, *Carbon*, 48, 107.
- [4] Zweben C., 2005, *Adv. Mater. Process*, 163, 33.
- [5] Zweben C., 2006, *Power Electron. Technol.*, 2, 40.
- [6] Lostetter, A., Barlow F., and Elshabini, A., 1998 *Adv. Microelectronics*, 25, 25.
- [7] Pintsuk G., Brunings S. E., Linke J., Smid J., and Xue L., 2003, *Fusion Eng. Design*, 66 – 68, 237.
- [8] Chu K., Guo H., Jia C., Yin F., Zhang X., Liang X., and Chen H., 2010, *Nanoscale Res. Lett.*, 5, 868.
- [9] Goyal V., and Balandin A. A., 2012, *Appl. Phys. Lett.*, 100, 073113.
- [10] Correia J. B., Livramento V., Shohoji, N., Tresso E., Yamamoto. K., Taguchi T., Hanada K., and Osawa, E., 2008, *Mater. Sci. Forum*, 744, 443.
- [11] Weeks J. R., and Sommer J. L., 1998, *US Patent* 5834115.
- [12] Moores K. A., and Joshi Y. K., 2001, *Future Circuits Int'l.*, 7, 45.